Spin-Wave Theory of Planar Ferromagnets and Antiferromagnets*

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The temperature dependence of the magnetization is calculated for a tetragonal crystal structure in which each atom has strong ferromagnetic coupling to the nearest neighbors in its own plane, but much weaker ferromagnetic or antiferromagnetic coupling to atoms in other planes. It is shown that, for a ferromagnet, the decrease of magnetization goes as $\tau^{3/2}$ for low temperatures and as $\tau \ln\tau$ for higher temperatures. The antiferromagnetic sublattice magnetization is shown to have the same temperature dependence as a two-dimensional ferromagnet with an effective anisotropy energy. The free energy is calculated in both cases and the results are used to show that the sublattice magnetization should be slightly larger on the antiferromagnetic side of an exchange-inversion transition.

RECENTLY, several crystals with a planar magnetic structure have been the subject of experimental investigation. The crystals are characterized by strong ferromagnetic coupling of each atom to neighboring atoms in the same plane and a much weaker coupling, either ferromagnetic or antiferromagnetic, to atoms in other planes. Examples are the hexagonal layer compounds $CrBr_3$ and $CrCl_3$, which are ferromagnetic[1] and antiferromagnetic,[2] respectively, at low temperatures. Another example is chromium-modified[3,4] Mn_2Sb which exhibits a first-order exchange inversion transition, from a ferrimagnetic to an antiferromagnetic state, with decreasing temperature.

The purpose of the present paper is to calculate the temperature dependence of the magnetization and the free energy in the type of structure described above, using spin-wave theory. The calculations are done for temperatures which are small compared to the Curie temperature (or, equivalently, to the intraplanar exchange interaction) but which can be large compared to the interaction between planes. The free energy expressions are used to study Kittel's model of the exchange-inversion transition[5] in order to compare the sublattice magnetizations on opposite sides of the transition.

FERROMAGNETIC INTERACTION

The model we shall use is a tetragonal magnetic lattice with strong ferromagnetic coupling, J_t, to the four nearest neighbors in the (001) plane, but with much weaker coupling, J_l, to the two nearest neighbors in adjacent planes. The nearest neighbor distances in the transverse and longitudinal directions are, respectively, a_t and a_l. The results can easily be extended to other planar structures.

The Hamiltonian is written in the Heisenberg form:

$$\mathcal{H}=-\sum_{j,\delta} J_\delta \mathbf{S}_j\cdot\mathbf{S}_{j+\delta}-2\mu_0 H_A\sum_j S_{jz}. \tag{1}$$

The sum j is over the N atoms of the lattice, while the sum $\boldsymbol{\delta}$ is over nearest neighbors; H_A is the effective anisotropy field. We write the Hamiltonian to bilinear terms in spin-wave operators as

$$\begin{aligned}&\mathcal{H}=-NS^2\sum_\delta J_\delta-2\mu_0 H_A NS+\mathcal{H}_0;\\ &\mathcal{H}_0=2S\sum_{\mathbf{k}}\sum_\delta J_\delta(1-\cos\mathbf{k}\cdot\boldsymbol{\delta})b_{\mathbf{k}}^\dagger b_{\mathbf{k}}+2\mu_0 H_A\sum_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}}.\end{aligned} \tag{2}$$

From (2) the magnon dispersion relation is

$$\omega_{\mathbf{k}}=2S\sum_\delta J_\delta(1-\cos\mathbf{k}\cdot\boldsymbol{\delta})+2\mu_0 H_A. \tag{3}$$

For small k_x, k_y, k_z, surfaces of constant $\omega_{\mathbf{k}}$ will be ellipsoids with the major axis in the k_z direction. The zone boundaries at $k_z=\pm\pi/a_l$ will be encountered at much lower values of $\omega_{\mathbf{k}}$ than the boundaries along the other axes. An appropriate expansion of $\omega_{\mathbf{k}}$ is then

$$\omega_{\mathbf{k}}=2SJ_t k_t^2+4SJ_l(1-\cos k_z)+2\mu_0 H_A, \tag{4}$$

where $k_t^2=k_x^2+k_y^2$. In this relation we have eliminated the lattice coordinates a_t, a_l by making the coordinate transformation $a_t k_x$, $a_t k_y$, $a_l k_z\rightarrow k_x$, k_y, k_z.

Using $M_s=2S\mu_0/a_t^2 a_l$ as the saturation magnetization of a simple tetragonal lattice, we have

$$\frac{M_s-M_F(\tau)}{M_s}\equiv\frac{\Delta M_F(\tau)}{M_s}=\frac{1}{(2\pi)^3 S}\times\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}dk_x dk_y dk_z\frac{1}{e^{\omega_{\mathbf{k}}/\tau}-1}. \tag{5}$$

For $\tau<SJ_t$ this integral will be uneffected if the limits of the k_x, k_y integrations are extended to infinity. Then

$$\frac{\Delta M_F}{M_s}=\frac{1}{(2\pi)^2 S}\left(\frac{\tau}{2SJ_t}\right)\int_0^\pi dk_z\times\ln\left\{1-\exp\left[-\frac{4SJ_l}{\tau}(1-\cos k_z)-\frac{2\mu_0 H_A}{\tau}\right]\right\}. \tag{6}$$

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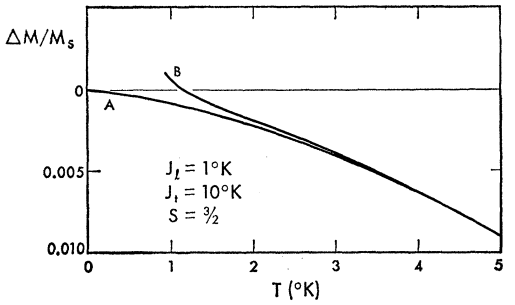

FIG. 1. Comparison of exact expression for $\Delta M_F/M_s$ with high-temperature expansion. Curve A, numerical evaluation of $\Delta M_F/M_s$. Curve B, first three terms in high-temperature expansion of $\Delta M_F/M_s$.

For $H_A=0$, (6) can be expanded in powers of SJ_l/τ by noting that

$$\ln(1-e^{-x})=\ln x-\frac{x}{2}+\frac{x^2}{24}+\cdots.$$

Upon expanding the integrand of (6) in this manner, and then performing the integration between the limits shown, the following expansion for the magnetization is obtained:

$$M_F(\tau)=M_s\Big[1-\frac{1}{4\pi S}\frac{\tau}{2SJ_t}\times\ln\frac{\tau}{2SJ_l}-\frac{J_l}{4\pi SJ_t}+\frac{J_l^2}{8\pi J_t\tau}+O(\tau^{-2})\Big]. \quad (7)$$

This is the expected form of the magnetization for $SJ_l\ll\tau<SJ_t$.

The leading terms in this expansion do not give a good approximation to (6) if $\tau\leqq SJ_l$. In that region one can extend the upper limit of the k_z integral in (6) to infinity and obtain an expression similar to Dyson's expansion in half-integral powers of τ[6]:

$$M_F(\tau)=M_s\Big\{1-\frac{1}{8\pi^{3/2}S}\frac{\tau^{3/2}}{(2S)^{3/2}J_tJ_l^{1/2}}\zeta(\tfrac{3}{2})-\frac{1}{192\pi^{3/2}S}\times\frac{\tau^{5/2}}{(2S)^{5/2}J_tJ_l^{3/2}}\Big[3\frac{J_l}{J_t}+\frac{3}{2}\Big]\zeta(\tfrac{5}{2})+O(\tau^{7/2})\Big\}; \quad (8)$$

here $\zeta(z)$ is the Riemann zeta function. Equation (8) is analogous to the expression of Gossard *et al.* for $CrBr_3$.[1]

Figures 1 and 2 compare the results of a numerical evaluation of (6) with the two expressions (7) and (8), respectively. It is seen that the expansion (8) is valid only for $\tau\leqq 8SJ_l$. The change of $\Delta M_F(\tau)$ from a $\tau^{3/2}$ law to a $\tau\ln\tau$ law for large τ is analogous to the transition of the specific heat of graphite from a τ^3 to a τ^2 law with increasing temperature.[7] For graphite the small elastic stiffness constant between adjacent hexagonal layers produces a phonon dispersion relation analogous to Eq. (4).

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ANTIFERROMAGNETIC INTERACTION

Our model is the same as that of the preceding section, with the exception that the interplanar coupling J_l now favors antiferromagnetism. The Hamiltonian is

$$\mathcal{H}=-J_t\sum_{j,\delta_t}\mathbf{S}_j\cdot\mathbf{S}_{j+\delta_t}+J_l\sum_{j,\delta_t}\mathbf{S}_j\cdot\mathbf{S}_{j+\delta_t}-2\mu_0H_A\sum_j S_{jz}{}^a+2\mu_0H_A\sum_j S_{jz}{}^b. \quad (9)$$

The coupling within each lattice plane, J_t, is ferromagnetic. The antiferromagnetic coupling between planes produces a ground state consisting of two sublattices with spins directed in opposite directions; the two planes adjacent to any plane of one of the sublattices belong to the opposite sublattice. In (9) the spins of sublattice a point in the $+z$ direction.

The methods of Anderson[8] or Kubo[9] enable us to reduce the Hamiltonian (9) to a spin-wave Hamiltonian. To bilinear terms in spin-wave operators we have

$$\mathcal{H}=-NS^2\textstyle\sum_\delta|J_\delta|-2N\mu_0H_AS+\mathcal{H}_0;$$
$$\mathcal{H}_0=\textstyle\sum_{\mathbf{k}}\omega_{\mathbf{k}}(b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}+c_{\mathbf{k}}{}^\dagger c_{\mathbf{k}})-\sum_{\mathbf{k}}(\omega_0-\omega_{\mathbf{k}}). \quad (10)$$

There are two spin-wave states for each $\mathbf{k}$ as indicated by the two sets of occupation numbers. The second summation over $\mathbf{k}$ gives the zero-point lowering of the energy. The magnon spectrum, $\omega_{\mathbf{k}}$, is given by

$$\omega_{\mathbf{k}}=(\omega_0{}^2-\omega_1{}^2)^{1/2}; \quad (11)$$

here

$$\omega_0=4J_lS+4SJ_t(2-\cos k_x-\cos k_y)+2\mu_0H_A;$$
$$\omega_1=4J_lS\cos(k_z/2). \quad (12)$$

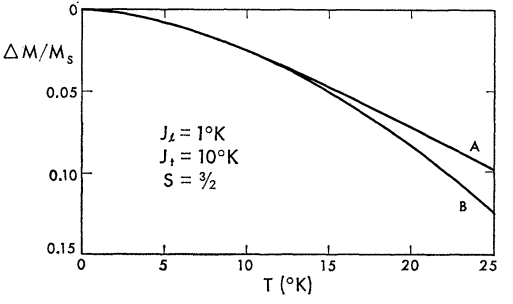

FIG. 2. Comparison of exact expression for $\Delta M_F/M_s$ with low-temperature expansion. Curve A, numerical evaluation of $\Delta M_F/M_s$. Curve B, $\tau^{3/2}$ and $\tau^{5/2}$ terms in low-temperature expansion of $\Delta M_F/M_s$.

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[9] R. Kubo, Phys. Rev. **87**, 568 (1952).

In these relations we have again transformed to dimensionless wave vectors $\mathbf{k}$.

For a cubic lattice, the temperature dependence of the sublattice magnetization is given by

$$\frac{\Delta M_A(\tau)}{M_s}=\frac{1}{S(2\pi)^3}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}dk_x dk_y dk_z \times\left\{\frac{1}{2}\left(\frac{\omega_0}{\omega_k}-1\right)+\frac{\omega_0/\omega_k}{e^{\omega_k/\tau}-1}\right\}. \quad (13)$$

The first term in the integrand is the zero-point lowering of the sublattice magnetization. We again set $H_A=0$. Since $J_l\ll J_t$, ω_k can be expanded in the following series:

$$\omega_k=2SJ_t k_t^2+4SJ_l -4SJ_l\left(\frac{J_l}{J_t k_t^2}\right)\cos^2\left(\frac{k_z}{2}\right)+O\left(\frac{J_l}{J_t k_t^2}\right)^2. \quad (14)$$

We see that $\omega_k\approx\omega_0$ except for a small region around $k_t=0$. For J_l/J_t sufficiently small we can neglect the remaining terms in ω_k and take $\omega_k=\omega_0$. In this approximation all the zero-point effects vanish and the calculation of $M_A(\tau)$ is greatly simplified. As a specific example, for $J_l/J_t=0.1$, the zero-point decrease in sublattice magnetization is only $0.003/S$, as compared with Anderson's value of $0.078/S$ for a completely antiferromagnetic cubic lattice.[8]

In the approximation $\omega_k\approx\omega_0$, Eq. (13) may be integrated exactly:

$$M_A(\tau)=M_s\left\{1+\frac{1}{4\pi S}\left(\frac{\tau}{2SJ_t}\right)\ln[1-\exp(-4SJ_l/\tau)]\right\}. \quad (15)$$

The magnetization curve of $CrCl_3$ appears to be fit very well by an expression of this form.[2] Our result has the same form as that of a two-dimensional ferromagnet with an anisotropy energy of $4SJ_l$.

FREE ENERGY AND APPLICATION TO EXCHANGE-INVERSION TRANSITIONS

The free energies of the ferromagnetic and antiferromagnetic lattices are evaluated from the expression $F=-\tau\ln Z$. Using the appropriate boson partition function Z, the free energy per atom at $H_A=0$ is given by

$$\frac{F(\tau)}{N}=-4J_tS^2-2J_lS^2-\frac{\tau}{(2\pi)^3}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}dk_x dk_y dk_z \times\ln[1-\exp(-\omega_k/\tau)]. \quad (16)$$

This expression is exact for the ferromagnetic problem and holds for the antiferromagnetic case in the limit $\omega_k\approx\omega_0$. We evaluate (16), using for ω_k first Eq. (4) with $H_A=0$, and then the first two terms of (14). The results in the two cases are, for $SJ_l\ll\tau<SJ_t$:

$$\frac{F_F(\tau)}{N}=-4J_tS^2-2J_lS^2 -\frac{\pi\tau^2}{48SJ_t}+\frac{\tau J_l}{2\pi J_t}\ln\frac{\tau}{2SJ_l}+\cdots, \quad (17)$$

$$\frac{F_A(\tau)}{N}=-4J_tS^2-2J_lS^2 -\frac{\pi\tau^2}{48SJ_t}+\frac{\tau J_l}{2\pi J_t}\left(1+\ln\frac{\tau}{4SJ_l}\right)+\cdots. \quad (18)$$

The free energies (17) and (18) can be applied to Kittel's model of the exchange-inversion transition.[5] In that model, the interaction between planes is assumed to be of the form $J_l=-\rho(a-a_c)$, where a is the spacing between planes and a_c is the spacing at which the interaction would change from ferromagnetic to antiferromagnetic. The free energy of the entire system is

$$F=\tfrac{1}{2}RV(a-a_T)^2+F_{\text{mag}}. \quad (19)$$

The first term of (19) is the free energy of the lattice; R is an elastic stiffness constant, a_T is the equilibrium spacing between planes in the absence of the magnetic interactions, and V is the volume. The temperature dependence of the free energy enters primarily through the temperature dependence of a_T. Kittel takes $F_{\text{mag}}=\mp\rho'(a-a_c)VM^2$, where the upper sign corresponds to the ferromagnetic state and the lower to the antiferromagnetic state.[10] M is the sublattice magnetization. The equilibrium values of a in the two states, a_F and a_A, are found by setting $\partial F/\partial a=0$. The transition from one state to the other will occur when $F_{\text{ferr}}(\tau)-F_{\text{antif}}(\tau)$ goes through zero. Using the form of F_{mag} mentioned above, Kittel finds that the transition occurs at a temperature such that $a_T(\tau)=a_c$. Furthermore, the equilibrium values of a at this temperature obey the relation

$$a_F-a_c=a_c-a_A. \quad (20)$$

One might expect that, if the temperature-dependent functions (17) and (18) are used for F_{mag} in Eq. (19), the simple results quoted above would no longer hold. Upon carrying out the steps described above with the new form of F_{mag}, it is found that the condition for a transition to occur, $a_T=a_c$, is replaced by

$$a_T(\tau)=a_c-\left(\frac{N}{V}\right)\frac{\rho\tau}{4\pi RJ_t}(1-\ln 2). \quad (21)$$

However, it is found that the lattice spacing on opposite sides of the transition still obeys Eq. (20). This relation

[10] The parameter ρ' differs from the ρ introduced earlier by dimensional factors, so that F_{mag} will have the dimensions of an energy.

states that the longitudinal coupling, J_l, changes sign during the transition, but its magnitude is the same on both sides.

Equations (7) and (15) can then be compared directly at the transition temperature:

$$\frac{M_A-M_F}{M_s}=\frac{\tau \ln 2}{8\pi S^2 J_t}. \tag{22}$$

The sublattice magnetization in the antiferromagnetic state is expected to be slightly larger than the magnetization in the ferromagnetic state. The quantity on the right side of (22) would be of the order of 10^{-2} for reasonable choices of the parameters.

Note added in proof. A. Yoshimori [Phys. Rev. **130**, 1312 (1963)] has derived similar results for antiferromagnetic crystals using a semicontinuum model for the ferromagnetic interaction. A. Narath [Phys. Rev. **131**, 1929 (1963)], has extended the measurements on $CrCl_3$ to below 1°K and has found that the approximation $\omega_k=\omega_0$ introduces errors in the predicted magnetization curve that are larger than the experimental uncertainties.

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Satellites in the Magnetoplasma Resonance in Sodium*

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The low-frequency magnetoplasma resonance ("whistler" or "helicon") has been studied in rectangular parallelepiped samples of Na at 4°K using improved measuring techniques and higher magnetic fields in order to obtain greater resolution. Structure in the resonance was observed as satellites of the previously reported resonances. The observed frequencies of these satellites fit the simple formula reported earlier.

INTRODUCTION

THE magnetoplasma resonance considered in this paper is a dynamic form of the Hall effect, which was first observed in Na at 4°K.[1] The effect can be observed as a standing, circularly polarized electromagnetic wave in a pure metal sample.[1–4] A detailed study of the frequencies of the resonance in many parallelepiped samples was made by Rose, Taylor, and Bowers.[2] This paper is an extension of that work with higher resolution which has brought to light the existence of satellite structure on the resonances observed previously.

Rose *et al.*[2] considered rectangular parallelepiped samples with sides X, Y, and Z placed in a large magnetic field along the z direction. It was assumed that the standing wave pattern in the sample would have an integral number of half-wavelengths in each of the three directions. A series of resonances was observed corresponding to modes with a single half-wavelength in each of the x and y directions, but a varying number of half-wavelengths in the z, or field, direction. Other investigators[3,4] have also reported modes with varying numbers of half-wavelengths along the magnetic field. However, no modes with more than one half-wavelength in either of the directions transverse to the magnetic field have been reported. We shall call such modes "transverse modes." The present work observes and examines the resonant peaks which arise from these transverse modes. These appear as satellites of those peaks observed by previous investigators.

The increased resolution which made this work possible was due to two new features in the instrumentation. First, phase-sensitive detection increased the signal to noise ratio by an order of magnitude[5]; and second, the use of a magnetic field of 27.4 kG produced resonances having a Q of from 20 to 30 in the very pure Na ($\rho_{300°K}/\rho_{4°K}\approx 7500$). This Q is 3 to 4 times greater than the highest Q's previously reported for the magnetoplasma effect.

THEORY

The dispersion relation for the magnetoplasma wave propagating in an unbounded metal (neglecting elec-

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